

## Temperature-Independent Aggregation Behavior of Polymer Donors Enabling Processing Condition-Tolerant, Additive-Free High-Performance All-Polymer Solar Cells

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In this work, the impact of temperature-dependent aggregation (TDA) behaviors in benzodithiophene (BDT)-based polymer donors ( $P_D$ s) on the electrical, morphological and photovoltaic performances of all-polymer solar cells (all-PSCs) is investigated. Two  $P_D$ s, PBDB-T and PBDB-Bz, with thienyl or benzylthienyl side chains and identical backbones are prepared. In contrast to PBDB-T, PBDB-Bz with bulkier side chains exhibits strong aggregation behavior in solution independent of temperatures between 20 and 100 °C. Notably, PBDB-Bz-based all-PSCs show a power conversion efficiency (PCE) of over 9% without any solvent additives (SA) or thermal annealing (TA), whereas these treatments are inevitable in optimizing the PCE of PBDB-T-based all-PSCs. Furthermore, high PCEs for the PBDB-Bz-based devices are maintained irrespective of their processing temperatures ( $T_{proc}$ ). However, the PCEs of PBDB-T-based devices are strongly dependent on their SA, TA and  $T_{proc}$  conditions. Thus, the development of  $P_D$ s with temperature-insensitive, strong aggregation behavior is crucial in producing reproducible,  $T_{proc}$ -tolerant and additive-free high-performance all-PSC devices.